

## EFFICIENCY CALIBRATION OF SOLID STATE ALPHA SPECTROMETERS

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Solid state alpha spectrometers have efficiency calibration requirements that differ significantly from most other types of radiation detectors, such as gamma spectrometers, gas proportional counters and liquid scintillation counters. The vast majority of samples analyzed by alpha spectroscopy utilize isotopic dilution in which an alpha emitting tracer is added during the preparation. In these cases, correct analytical results do not depend on the detector calibration and the issue is important only for purposes of accurately determining the chemical yield of the analysis, which is useful in assessing the overall performance of the method. Only in the minority of cases in which no alpha tracer is added, such as the analysis of  $^{237}\text{Np}$  using beta emitting  $^{239}\text{Np}$  as a tracer, does the proper determination of detector efficiency directly affect the accuracy of the results. It is for these analyses that extreme care must be taken to properly calibrate the detector system.

Proper calibration consists of two components: Procurement of appropriate calibration standards and proper utilization of these standards.

The following discussions assume that routine samples are electroplated on metal planchets for counting.

Calibration standards must meet the following criteria to be suitable alpha spectroscopy efficiency standards:

1. They must be prepared in the exact same geometry as the actual samples to be counted. This means that they must be plated over the same area on plates of the same thickness and diameter.
2. They must have traceability (preferably to NIST) that can be easily documented.
3. They must either be produced and/or calibrated by NIST directly, or be obtained from a facility that has successfully passed a QA audit by the user laboratory.
4. They must have traceability re-certified on a periodic basis.
5. They must be physically robust enough to endure normal handling in the lab over an extended period without losing activity.

## **PURCHASING STANDARDS**

Purchasing standards from a commercial supplier of radioactivity standards is an option, but compliance with all the above requirements can be difficult and costly. Duplicating the geometry exactly is the first hurdle. The diameter of the plated area on the plate is the most critical issue, particularly when counting samples very close to the surface of the detector to maximize sensitivity. If finding the correct size commercially is a problem, the lab can prepare the plate and send it to a commercial source for calibration only. In this case stabilization of the activity on the plate is important to eliminate loss of activity during shipping.

The adequacy of the documentation of traceability of a purchased standard is the decision of the lab's QA department, which in turn has to defend it to external auditors. This documentation can range from a simple statement on the certificate that it is traceable, to an actual certificate from NIST.

The issue of QA approval of suppliers is becoming an increasingly expensive one. To fully comply with QA programs meeting 10CFR50, etc., a lab must audit a supplier of quality related services, unless the supplier is NIST.

Re-certification is more of an expense issue, more than a technical issue. It is assumed that any commercial facility (or NIST) producing or calibrating a standard would be able to re-certify it on a periodic basis for a fee.

Most commercially purchased standards would have the activity fixed sufficiently to allow normal usage with little fear of loss of activity. Related to the first requirement concerning geometry, care must be taken to assure that the activity is not fixed in such a way that the scattering of the alphas is significantly greater than that for routine samples prepared by the lab. Some commercial standards have the activity embedded in metallic layers that, while very stable, render them unusable for alpha spectroscopy standards.

## **PREPARATION OF STANDARDS BY THE LAB**

It is difficult for a laboratory to internally prepare calibration standards for alpha spectroscopy because of the lack of a gravimetric determination of how much of a standard solution is deposited on a counting planchet. This discussion will cover five methods of preparing appropriate standards: 1. Electrodeposition followed by calibration on a traceable gross alpha counter. 2. Electrodeposition followed by calibration by an external calibration service. 3. Exhaustive electrodeposition. 4. Stippling a known amount of activity. 5. Dual plate electrodeposition with a tracer.

1. **ELECTRODEPOSITION FOLLOWED BY CALIBRATION ON A TRACEABLE GROSS ALPHA COUNTER** - This method would meet the requirement for identical geometry as the samples, assuming the standard is plated in the same manner. If the activity is plated on a noble metal, such as platinum, and flamed to redness afterward, the activity can be very stable. Physical stiffness can be enhanced by using a Pt-Rh

alloy, but the material costs are higher. Since it is not known how much activity is plated, the standard is then calibrated on a gross alpha counter that has separate and traceable calibration. This is obviously only a partial solution, since the gross counter still needs to be calibrated. A gross alpha counter may be somewhat more easily calibrated than an alpha spectrometer, however, because there may be less concern for the calibrating standard to have identical geometry as samples, especially for an internal  $2\pi$  flow counter. Backscatter is an issue with high geometry internal counters, and care should be taken to assure that the backing material for the standard is the same as for the plate being calibrated.

2. **ELECTRODEPOSITION FOLLOWED BY CALIBRATION BY AN EXTERNAL CALIBRATION SERVICE** - This method meets the requirements for geometry and stability as described in option 1, but incurs the costs of calibration service and auditing. NIST's current price for alpha plate calibration is over \$1400, which would recur periodically according to the recertification schedule. Other commercial sources are significantly less expensive, but the costs of on-site audits can be prohibitive. The audit costs can be amortized over other uses if the lab has already audited a source of radioactive solution standards and can obtain plate calibration service from the same source.
3. **STIPPLING** - A precisely known amount of activity can be carefully stippled onto a plate in a pattern that closely approximates the typical plating distribution. This method meets the geometry requirement as long as the pattern is carefully controlled. Unless the plate is flamed after stippling, the activity is only tenuously attached to the plate and can be rubbed off very easily. Even after flaming, small amounts of residue remain that can be rubbed off easily, and is the primary drawback for this method.
4. **EXHAUSTIVE ELECTRODEPOSITION** - In this method, a known amount of activity is added to a plating cell and electroplated onto an appropriate metal plate. The plating solution is then quantitatively transferred back to its original beaker and the plated source is removed. A second plate is inserted into the plating cell, the original plating solution is treated and returned to the cell and a second plating is performed. This process is repeated a third time, and then the residual solution is transferred to a planchet suitable for gross alpha counting and evaporated. All three plates are counted on the same detector (either an alpha spectrometer or a gross alpha counter) and the gross alpha planchet is counted (to assure that no significant amount of activity was left unplated on the three plates). Assuming that all transfers were quantitative, the fraction of the total activity that was plated on the first

plate can be calculated from all the counting data, and therefore the plate has a known amount of activity. This meets all requirements for geometry, stability and traceability and eliminates requirements for external audits and calibration service costs. The primary problems are that the method is tedious and it is difficult to control splattering when treating the residual plating solutions. These problems are eliminated in the next method.

5. DUAL PLATE ELECTRODEPOSITION WITH A TRACER - This method is similar to the exhaustive electrodeposition but eliminates many of its drawbacks. A known amount of activity is again added to a plating cell and electroplated onto an appropriate plate. A quantitative transfer of the residual plating solution back to its original beaker is then performed and the plate is removed and flamed. A known amount of alpha tracer, of the same element but different nuclide than the original activity added, is then added to the residual solution. This solution is then treated again and transferred back to the plating cell, and electroplated onto a second plate. Since tracer has been added, the treatment and second transfer are now not required to be completely quantitative. The second plate is counted and, using isotopic dilution, the total amount of activity left unplated from the first plating can be determined precisely, assuming only that the first transfer was quantitative. The amount of activity on the first plate is then calculated as the total added at the beginning minus the amount left in the plating solution. This method meets all the stated requirements and is quick and inexpensive. It does require two traceable sources of alpha activity of the same element (such as  $^{242}\text{Pu}$  and  $^{239}\text{Pu}$ ), which are usually available anyway in most laboratories. It can also be performed with a slight modification in which only one of the sources need be traceable, but the math is more complicated and the calibrated plate ends up with two peaks instead of one. It is a good idea to perform this procedure on at least three standards to provide assurance of the precision of the method.